

# Temperature effects in polarized low-energy electron scattering from solids and liquids

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The effects of temperature on the polarization of low-energy electrons scattered from surfaces of liquids, polycrystals, and single crystals are discussed. The polarization of the single scattering component of the intensity is shown to be independent of temperature, whereas that of the multiple scattering intensity is temperature dependent. Differences between polarizations observed for electron scattering from free atoms and from polycrystals and liquids are qualitatively explained and a quantitative theory is outlined. The implications for polarized low-energy electron diffraction from single-crystal surfaces are also discussed.

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## I. INTRODUCTION

Measurement of changes in polarization produced by scattering low-energy electrons from surfaces provides a new experimental probe of surface properties. A limited number of measurements on crystals<sup>1,2</sup> and polycrystals<sup>3,4</sup> have been reported and the dynamical theory of low-energy electron diffraction (LEED) has been extended to include polarization effects.<sup>5</sup> The scattering from polycrystals is similar to that observed for free atoms, whereas scattering from crystals is not, apparently due to multiple scattering. It is hoped that these differences will provide more stringent tests of LEED theory and allow more precise determination of surface parameters. However, detailed comparisons between experimental and theoretical LEED results are not yet available.

Most electron scattering devices have insufficient energy resolution to resolve energy losses due to phonon scattering which remove intensity from the truly elastic scattering and redistribute it throughout the Brillouin zone as a temperature dependent background. At present LEED theory only treats the truly elastic scattering and phonon effects cannot be easily included. Thus it is important to understand the ways in which the phonon scattering will affect the experimental results so that meaningful comparisons with theoretical results can be made. The analysis presented in this paper shows that polarization of the single scattering component of the intensity is temperature independent whereas that of the multiple scattering intensity is temperature dependent. At temperatures high enough so that multiphonon scattering dominates, the scattering from crystals and liquids becomes identical. Polarization effects are treated for this case by extending the model of Schilling and Webb.<sup>6</sup>

## II. ELECTRON-ATOM SCATTERING

Polarization effects due to the spin-orbit interaction in electron scattering from free atoms have been studied extensively.<sup>7</sup> The polarization along a fixed direction  $\hat{n}_i$  is

$$\mathbf{P}_i \equiv \frac{N_{\uparrow} - N_{\downarrow}}{N_{\uparrow} + N_{\downarrow}} \hat{n}_i \equiv \frac{I_{\uparrow} - I_{\downarrow}}{I_{\uparrow} + I_{\downarrow}} \hat{n}_i, \quad (1)$$

where  $N_{\uparrow}$  and  $N_{\downarrow}$  are respectively the number of electrons with spins parallel and anti-parallel to  $\hat{n}_i$ ;  $I_{\uparrow}$  and  $I_{\downarrow}$  are the respective currents.

If  $g(E, \theta)$  and  $f(E, \theta)$  are the complex differential cross sections for scattering with and without a spin reversal, the total differential scattering cross section for an incident electron beam with polarization  $\mathbf{P}_o$ , energy  $E$  and wave vector  $\mathbf{k}_o$ ,  $|\mathbf{k}_o| = \hbar(2mE)^{1/2}$ , scattered through angle  $\theta$  into scattered wave vector  $\mathbf{k}$  is given by

$$\frac{d\sigma(E, \theta, \mathbf{P}_o)}{d\Omega} = \frac{d\sigma_o(E, \theta)}{d\Omega} (1 + S(\theta) \mathbf{P}_o \cdot \hat{n}), \quad (2)$$

where

$$\frac{d\sigma_o}{d\Omega}(E, \theta) = |f|^2 + |g|^2, \quad S(\theta) = i \frac{fg^* - gf^*}{|f|^2 + |g|^2}$$

and

$$\hat{n} = \frac{\mathbf{k}_o \times \mathbf{k}}{|\mathbf{k}_o \times \mathbf{k}|}.$$

$d\sigma_o/d\Omega$  is the cross section for unpolarized scattering;  $S(\theta)$ , the Sherman function, contains information about polarization effects. The polarization  $\mathbf{P}$  after scattering is given by<sup>7</sup>

$$\mathbf{P} = \frac{\{\mathbf{P}_o \cdot \hat{n} + S(\theta)\}\hat{n} + T(\theta)\hat{n} \times (\mathbf{P}_o \times \hat{n}) + U(\theta)(\hat{n} \times \mathbf{P}_o)}{1 + \mathbf{P} \cdot \hat{n}S(\theta)}, \quad (3)$$

where

$$T(\theta) = \frac{|f|^2 - |g|^2}{|f|^2 + |g|^2} \text{ and } U(\theta) = \frac{fg^* + gf^*}{|f|^2 + |g|^2}$$

describe the rotation of the polarization;  $S^2 + T^2 + U^2 = 1$ .

Total differential cross sections have been measured for several atoms<sup>7</sup> and accurate calculations of  $f$  and  $g$  for  $E \geq 75$  eV have been made for most elements.<sup>8</sup> For atoms maximum polarization effects are observed when  $d\sigma_o/d\Omega$  is small

so that large polarizations are associated with small intensities.

### III. SCATTERING FROM CRYSTALS

This section reviews electron scattering from crystal surfaces including the thermal motions of the atoms and discusses how these motions affect the different components of the scattered intensity. Only the case of a crystal composed of identical atoms is considered.

#### A. Single scattering

For single scattering from a rigid lattice the intensity is<sup>9</sup>

$$I(\Delta\mathbf{k}, T=0) = \frac{d\sigma}{d\Omega} \mathcal{J}^0(\Delta\mathbf{k}, \alpha), \quad (4)$$

where  $\mathcal{J}^0(\Delta\mathbf{k}, \alpha)$  is the rigid-lattice interference function,  $\hbar\Delta\mathbf{k} = \hbar(\mathbf{k} - \mathbf{k}_0)$  is the momentum transfer, and  $\alpha$  describes the electron attenuation.  $\mathcal{J}^0(\Delta\mathbf{k}, \alpha)$  contains all the information about the crystal structure and has maxima whenever the Laue conditions are satisfied; e.g.,  $\Delta\mathbf{k} \cdot \mathbf{a}_i = 2\pi M_i$ ,  $i = 1, 2, 3$  where  $\mathbf{a}_i$  are the unit cell vectors and  $M_i$  are integers. This is illustrated in Fig. 1(a). On the other hand, all the polarization effects are contained in  $d\sigma/d\Omega$  so that  $S(\theta)$  for the single scattered intensity is independent of crystal structure.

The effects of thermal motions on the single scattering have been reviewed by Lagally.<sup>10</sup> The diffracted intensity becomes

$$\begin{aligned} \langle I(\Delta\mathbf{k}, T, \mathbf{P}_1) \rangle &= \frac{d\sigma}{d\Omega} \sum_{ij} \alpha_i \alpha_j \exp[i\Delta\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)] \\ &\times \exp[-\langle (\Delta\mathbf{k} \cdot \mathbf{u}_i)(\Delta\mathbf{k} \cdot \mathbf{u}_j) \rangle] \\ &\{1 + \langle (\Delta\mathbf{k} \cdot \mathbf{u}_i)(\Delta\mathbf{k} \cdot \mathbf{u}_j) \rangle + \exp[\langle (\Delta\mathbf{k} \cdot \mathbf{u}_i)(\Delta\mathbf{k} \cdot \mathbf{u}_j) \rangle] \\ &\quad - 1 - \langle (\Delta\mathbf{k} \cdot \mathbf{u}_i)(\Delta\mathbf{k} \cdot \mathbf{u}_j) \rangle\}, \quad (5) \end{aligned}$$

where  $\mathbf{u}_i$  are the instantaneous atomic displacements from equilibrium positions  $\mathbf{r}_i$  and  $\langle \rangle$  represents a thermal average. As for rigid lattice scattering, Eq. (4), all the polarization effects are contained in  $d\sigma/d\Omega$  and, using Eq. (1),

$$P = \frac{\langle I(+\mathbf{P}_1) \rangle - \langle I(-\mathbf{P}_1) \rangle}{\langle I(+\mathbf{P}_1) \rangle + \langle I(-\mathbf{P}_1) \rangle} = \frac{d\sigma(+)/d\Omega - d\sigma(-)/d\Omega}{d\sigma(+)/d\Omega + d\sigma(-)/d\Omega},$$

i.e., the polarization is independent of temperature for single scattering.

#### B. Zero-, one-, and multiphonon interference functions

The three terms inside the braces in Eq. (5) are called the zero-, one-, and multiphonon intensities:

$$\langle I(\Delta\mathbf{k}, T) \rangle = I^0(\Delta\mathbf{k}, T) + I^1(\Delta\mathbf{k}, T) + I^m(\Delta\mathbf{k}, T). \quad (6)$$

The zero-phonon intensity  $I^0(\Delta\mathbf{k}, T)$  removes intensity from the elastically scattered beams and the one- and multiphonon intensities redistribute it throughout the Brillouin zone as thermal diffuse scattering.

The zero phonon intensity is just the intensity from a rigid lattice reduced by the Debye-Waller factor:

$$e^{-2M} \equiv \exp[-\langle (\Delta\mathbf{k} \cdot \mathbf{u}_i)(\Delta\mathbf{k} \cdot \mathbf{u}_j) \rangle]. \quad (7)$$

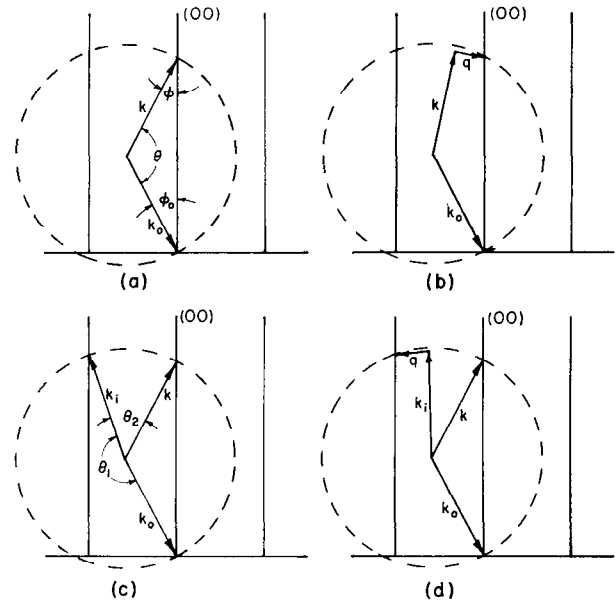


FIG. 1. (a) Single scattering at  $T = 0$  K; (b) single scattering at  $T > 0$  K including a one phonon scattering with wave vector  $\mathbf{q}$ ; (c) double scattering at  $T = 0$  K; (d) double scattering at  $T > 0$  K including a one phonon scattering with wave vector  $\mathbf{q}$ .  $\mathbf{k}_0$ ,  $\mathbf{k}_i$ , and  $\mathbf{k}$  are, respectively, the primary, intermediate and final wave vectors.

With no attenuation and  $\mathbf{u}_i$  independent of  $i$ ,  $2M = \langle (\Delta\mathbf{k} \cdot \mathbf{u})^2 \rangle$ , so that a determination of  $2M$  is a measure of the mean-square displacement along  $\Delta\mathbf{k}$ . In the high temperature limit,  $\langle u^2 \rangle$  is proportional to the temperature so that  $2M = 2M'T$ . Experimentally,  $2M'$  is determined from the slope of  $\ln \langle I \rangle$  versus  $T$ . Deviations from constant slope occur at low temperature due to the zero-point motion and at high temperature where the measurement includes diffuse scattering,  $I^1(\Delta\mathbf{k}, T)$  and  $I^m(\Delta\mathbf{k}, T)$ , whose properties are described below.

Expanding the displacements in Eq. (5) in terms of the normal modes of the lattice and taking the high temperature limit<sup>11</sup> yields for the one phonon intensity

$$I^{(1)}(\Delta\mathbf{k}, T) \propto \frac{d\sigma}{d\Omega} \frac{2Me^{-2M}}{|q^n|}, \quad (8)$$

where  $1 \leq n \leq 2$ .

The ratio of the integrated intensity in the one-phonon scattering to that in the zero-phonon scattering is  $2M$ .<sup>11</sup>

The remaining component in the diffuse scattering is the multiphonon scattering which is distributed uniformly throughout the zone.<sup>10</sup> The temperature dependence of the integrated multiphonon intensity from Eq. (5) is

$$\int I^m(\Delta\mathbf{k}) d\Delta\mathbf{k} \propto e^{-2M}(e^{2M} - 1 - 2M). \quad (9)$$

The temperature dependences of the integrated zero-, one-, and multiphonon intensities are shown in Fig. 2. For low-energy electron diffraction  $2M$  is typically greater than one so that less than one-third of the total scattering is in the zero-phonon component. At high temperatures nearly all the intensity is due to multiphonon scattering. The intensity at any point in the Brillouin zone can be written in the useful form<sup>11</sup>

$$I(\Delta\mathbf{k}) = e^{-2M} \mathcal{J}^0(\Delta\mathbf{k}) + 2Me^{-2M} \mathcal{J}^1(\Delta\mathbf{k}) + (1 - e^{-2M} - 2Me^{-2M}) \mathcal{J}^m(\Delta\mathbf{k}) \quad (10)$$

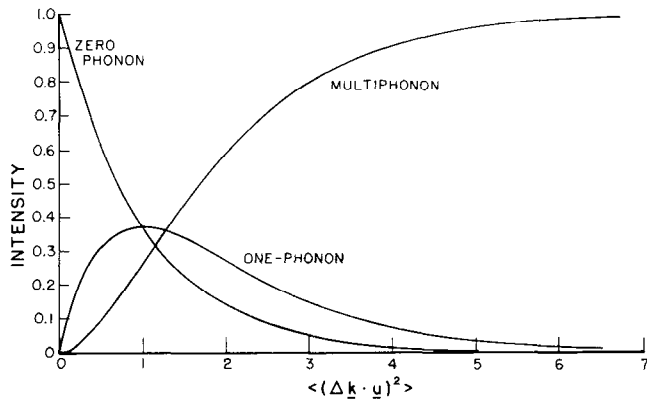


FIG. 2. Relative contributions of the zero-, one- and multiphonon scattering as a function of the exponent of the Debye-Waller factor  $\langle(\Delta\mathbf{k} \cdot \mathbf{u})^2\rangle$ , Eq. (7). In the high temperature limit,  $\langle(\Delta\mathbf{k} \cdot \mathbf{u})^2\rangle$  is directly proportional to the temperature. For most crystals  $\langle(\Delta\mathbf{k} \cdot \mathbf{u})^2\rangle$  is between one and two at room temperature and the three components contribute nearly equally to the total intensity.

where  $\mathcal{I}^0(\Delta\mathbf{k})$ ,  $\mathcal{I}^1(\Delta\mathbf{k})$ , and  $\mathcal{I}^m(\Delta\mathbf{k})$  are the shapes of the zero-, one-, and multiphonon interference functions respectively. These interference functions have been determined throughout the Brillouin zone for Ag(111)<sup>12</sup> and are expected to be similar for other materials; Fig. 3 shows results for the (666) reflection.

### C. Multiple scattering

Approximately one-half of the total elastic intensity arises from multiple scattering which dominates structure in intensity-energy profiles at low energies,  $E \lesssim 150$  eV. In addition, both theoretical calculations<sup>5</sup> and experiment<sup>1</sup> show that the polarization is significantly modified by multiple scattering; the change in polarization for each scattering is still determined by the scattering factor, but the net polarization produced by a series of scatterings also depends on the crystal structure which determines which multiple scattering sequences are important. Consider double scattering from a surface; for a rigid lattice, the Laue conditions are replaced by

$$\mathbf{k}_i^{\parallel} - \mathbf{k}_o^{\parallel} = \mathbf{G}_1^{\parallel} \text{ and } \mathbf{k}_i^{\parallel} - \mathbf{k}_i^{\parallel} = \mathbf{G}_2^{\parallel} \quad (11)$$

where  $\mathbf{k}_o^{\parallel}$ ,  $\mathbf{k}_i^{\parallel}$  and  $\mathbf{k}^{\parallel}$  are the surface components of the incident, intermediate and final wave vectors and  $\mathbf{G}_1^{\parallel}$  and  $\mathbf{G}_2^{\parallel}$  are surface reciprocal lattice vectors. Equation (11) is illustrated in Fig. 1(c). The incident beam direction and detector position fix both  $\mathbf{k}_o$  and  $\mathbf{k}$  for a given diffraction geometry and all possible  $\mathbf{k}_i$  are determined by Eq. (11).

At finite temperatures, Eq. (11) becomes

$$\mathbf{k}_i^{\parallel} + \sum_n \mathbf{q}_n^{\parallel} = \mathbf{G}_1^{\parallel} + \mathbf{k}_o^{\parallel} \text{ and } \mathbf{k}_i^{\parallel} - \sum_n \mathbf{q}_n^{\parallel} = \mathbf{k}^{\parallel} - \mathbf{G}_2^{\parallel} \quad (12)$$

and  $\mathbf{k}_i$  is defined only to within a sum of phonon wave vectors  $\sum_n \mathbf{q}_n^{\parallel}$ ; Fig. 1(d) gives an example for  $n = 1$ . All  $\mathbf{k}_i$  within the Brillouin zone can thus contribute to the multiple scattering. Because the zero-, one- and multiphonon scatterings each weight different regions of the Brillouin zone, Fig. 3, each will emphasize different  $\mathbf{k}_i$  and thus different scattering angles. This will be particularly important when the polarization is changing rapidly with scattering angle and will result in a

different total polarization for each thermal scattering component of the intensity. Additionally, the relative contribution of each thermal component to the total intensity has a different temperature dependence, Fig. 2 and Eq. (10), resulting in a temperature dependent total polarization. These temperature variations may be particularly important in LEED since the largest polarizations are observed near intensity minima<sup>1</sup> where the contribution from thermal diffuse scattering can dominate the intensity.<sup>13,14</sup> Preliminary measurements for W(100)<sup>1</sup> and Au(110)<sup>2</sup> show changes in polarization that may be due to temperature. Careful experimental work is needed to determine the magnitude of these effects since it will be difficult to quantitatively include thermal diffuse scattering in theoretical treatments.

For temperatures sufficiently large ( $2M \gtrsim 4$  in Fig. 3) that the multiphonon scattering dominates, the thermal vibrations are so large that crystal structure effects are no longer important. In this case, the theory for electron-liquid scattering described in Sec. V becomes appropriate and the polarization becomes similar to free atom polarization and is temperature independent.

### IV. SCATTERING FROM POLYCRYSTALS

The scattered intensity for polycrystals for fixed  $\mathbf{k}_o$ ,  $\mathbf{k}$  is obtained by integrating the results for each surface structure

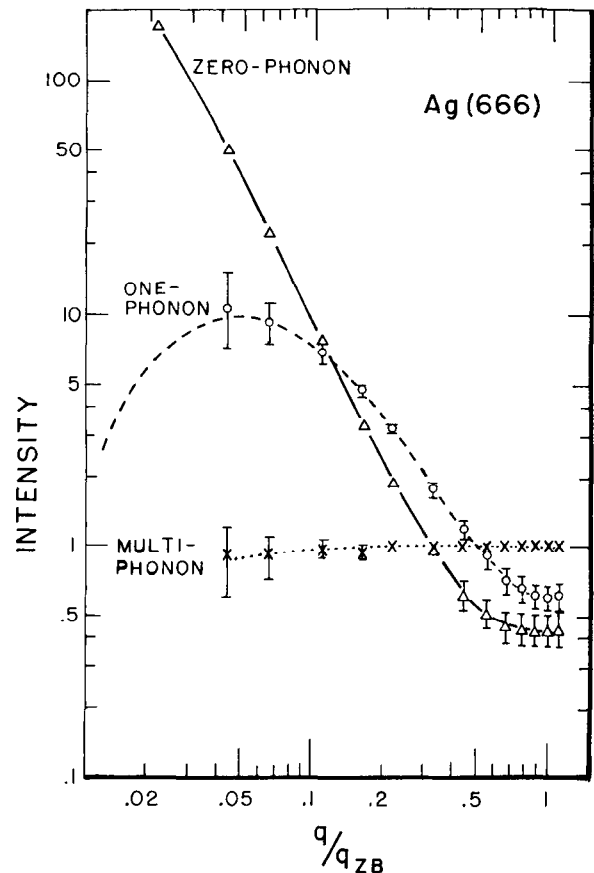


FIG. 3. Variation of the zero-, one-, and multiphonon scattering as a function of position in the Brillouin zone for the Ag(666) reflection;  $q_{ZB}$  is the wave vector reaching to the Brillouin zone boundary. (after Ref. 12); The one-phonon intensity must go to zero at the center of the zone since for a finite crystal there are no phonons with  $q = 0$ .

over all possible angles of incidence then summing over all the structures exposed on the surface weighting each according to the fractional area covered. This greatly increases the number of multiple scattering combinations for each  $\mathbf{k}_0$ ,  $\mathbf{k}$  so that the polarization of the multiply scattered intensity will be substantially reduced, leaving only the polarization due to single scattering. This will be particularly true at high temperature where  $I^m(\Delta\mathbf{k}, T)$  dominates and the scattering should be very similar to that for liquids discussed in the following section.

These considerations are consistent with experiment. The polarization of electrons scattered from solid Hg at 77 K<sup>3</sup> has an energy and angular dependence like that of a free Hg atom<sup>7</sup> but is reduced in magnitude. Within experimental error, the polarization from polycrystalline W is the same at 1300° C and 1700° C.<sup>4</sup>

## V. SCATTERING FROM LIQUIDS

A liquid (or amorphous solid) possesses only short range order. As a result, interference effects between atoms are even less important for a liquid than for a solid near the melting point where the multiphonon scattering dominates. We describe electron scattering from liquids by extending the model of Schilling and Webb<sup>6</sup> to include polarization effects.

In this model, it is assumed that: (1) the elastic scattering is described by the scattering cross section  $d\sigma/d\Omega$  which is known; (2) the linear attenuation coefficient is given by

$$\mu_T = \rho(\sigma_{in} + \sigma_{el})$$

where  $\rho$  is the number density of the atoms and  $\sigma_{in}$  and  $\sigma_{el}$  are the total inelastic and elastic cross-sections; (3) for  $\Delta k \gtrsim 5 \text{ \AA}^{-1}$  the interference function is constant so that it is appropriate to add intensities instead of amplitudes. Thus

$$I(\Delta\mathbf{k}, \mathbf{P}_1) = I_1(\Delta\mathbf{k}, \mathbf{P}_1) + I_2(\Delta\mathbf{k}, \mathbf{P}_1) + \dots, \quad (13)$$

where  $I_1(\Delta\mathbf{k}, \mathbf{P}_1)$  is the elastic intensity from single scattering given by

$$I_1(\Delta\mathbf{k}, \mathbf{P}_1) = \frac{\rho[d\sigma(E, \theta, \mathbf{P}_1)/d\Omega]}{\mu_T [1 + \cos\phi_0/\cos\phi]}, \quad (14)$$

where  $\phi_0$  and  $\phi$  are respectively the angles of incidence and exit, Fig. 1(a). Similarly, the twice scattered intensity is<sup>6</sup>

$$\begin{aligned} I_2(\Delta\mathbf{k}, \mathbf{P}_1) = & \int_0^\infty \exp(-\mu_T z_1 \sec\phi_0) \\ & \times \int_{4\pi} \rho \sec\phi_0 dz_1 \frac{d\sigma_0(\theta_1, E, \mathbf{P}_1)}{d\Omega} [1 + S(\theta_1)\mathbf{P}_1 \cdot \hat{n}_1] d\Omega_1 \\ & \times \int_{z_{\max}}^{z_{\min}} \exp(-\mu_T |z_2 - z_1| \sec\gamma) \\ & \times \rho \sec\gamma \frac{d\sigma_0(\theta_2, E, \mathbf{P}_2)}{d\Omega} [1 + S(\theta_2)\mathbf{P}_2 \cdot \hat{n}_2] dz_2, \quad (15) \end{aligned}$$

where  $\theta_1$  and  $\theta_2$  are the scattering angles for the first and second scatterings respectively and are related to  $\theta$  by

$$\cos\theta_2 = \cos\theta \cos\theta_1 + \sin\theta \cos\theta_1 \sin\delta \quad (16)$$

with  $\delta$  the angle between the planes defined by  $(\mathbf{k}_0, \mathbf{k})$  and  $(\mathbf{k}_0, \mathbf{k}_1)$ ;  $\hat{n}_1 \equiv (\mathbf{k}_0 \times \mathbf{k}_1)/|\mathbf{k}_0 \times \mathbf{k}_1|$  and  $\hat{n}_2 \equiv (\mathbf{k}_1 \times \mathbf{k})/|\mathbf{k}_1 \times \mathbf{k}|$ ;  $\mathbf{P}_1$  is the incident polarization and  $\mathbf{P}_2$  is the polarization after

the first scattering obtained from Eq. (3),  $\gamma$  is the angle between  $\mathbf{k}_1$  and the surface normal;  $z_1$  and  $z_2$  are the depths of the first and second scatterings. Similar terms results for higher order processes; each successively higher term contains an additional factor  $\mu_T^{-1} |d\sigma(\theta_i, E, \mathbf{P}_i)/d\Omega|$ . The series, Eq. (13), converges rapidly since  $\sigma_{in} \gtrsim \sigma_{el}$ ; physically, for an electron to be multiply scattered it must travel further through the material and will thus be more strongly attenuated. This model gives good results for unpolarized scattering from both liquids<sup>6</sup> and solids at high temperatures.<sup>9</sup>

Equation (15) is effectively a self-convolution of  $d\sigma/d\Omega$  under the constraint Eq. (16)<sup>6</sup> and is dominated by the forward scattering. Since the scattering is strong in the forward direction and  $S(\theta)$  very small, the polarization arising from Eq. (15) is also expected to be very small. In addition to multiple scattering contributions  $I_M$  are found to be nearly independent of  $\theta^6$  so that Eq. (13) can be written

$$I(\Delta\mathbf{k}, \mathbf{P}_1) = I_1(\Delta\mathbf{k}, \mathbf{P}_1) + I_M.$$

The measured Sherman function is given by

$$S_{\text{liquid}} = S_{\text{atom}} [1 + I_M/I_S]^{-1} \quad (17)$$

when

$$I_S = \frac{1}{2} [I_1(+\mathbf{P}_1) + I_1(-\mathbf{P}_1)].$$

Since  $I_M$  and  $I_1$  are positive,  $S_{\text{liquid}}$  will have the same angular and energy variations as  $S_{\text{atom}}$  but will be reduced in magnitude.

There are no measurements of the polarization for electron-liquid scattering. But, as discussed in Sec. IV, Eq. (17) is consistent with observations for polycrystals.<sup>3,4</sup>

## VI. CONCLUSIONS

For low-energy electron scattering from single-crystal surfaces, the polarization of the single scattering is found to be independent of temperature whereas that of the multiple scattering is temperature dependent. The differences between polarizations observed for electron scattering from free atoms and liquids are due to the unpolarized multiple scattering background in liquid scattering.

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